

# Cotunneling and one-dimensional localization in individual disordered single-wall carbon nanotubes: Temperature dependence of the intrinsic resistance

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We report on the temperature dependence of the intrinsic resistance of long individual disordered single-wall carbon nanotubes. The resistance grows dramatically as the temperature is reduced, and the functional form is consistent with an activated behavior. These results are described by a Coulomb blockade along a series of quantum dots. We occasionally observe a kink in the activated behavior that reflects the change of the activation energy as the temperature range is changed. This is attributed to charge hopping events between nonadjacent quantum dots, which is possible through cotunneling processes.

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## I. INTRODUCTION

Single-wall carbon nanotubes (SWNT's) are an excellent system to study one-dimensional (1D) transport. In particular, the effect of disorder in 1D is very pronounced; current lines have to follow the wire and cannot go round impurity centers. As the transmission of impurity centers becomes low enough, the 1D wire is divided into a series of quantum dots. The conduction is then thermally activated  $R(T) \approx \exp(T^{-1})$ .<sup>1-4</sup>

Measurements of 2D or 3D arrays of quantum dots can show a slower than thermally activated dependence of the conduction  $R(T) \approx \exp(T^{-0.5})$ .<sup>5,6</sup> This has been recently attributed to cotunneling processes, which allow charge transfer between nonadjacent quantum dots.<sup>7-9</sup> Indeed, cotunneling transport in a series of quantum dots is analogous to variable-range hopping (VRH).<sup>10</sup> Charges try to find hopping events with the lowest activation energy and the shortest hopping distance. The slower than thermally activated dependence of the conduction is then a result of successive thermally activated curves with an activation energy that decreases as the temperature is reduced. However, such a succession of activated curves remains to be observed.

Localization experiments have been carried on nanotube films or individual SWNT's contacted to microfabricated electrodes, but tube-tube junctions and tube-electrode interfaces make the analysis difficult.<sup>4,11-14</sup> In our experiments, the intrinsic resistance of disordered SWNT's is measured in a four-point configuration.<sup>15</sup> The intrinsic resistance is found to be thermally activated. As the gate voltage ( $V_g$ ) is swept, we observe Coulomb blockade oscillations that can be rather regular in some cases. These measurements are consistent with a series of quantum dots that are typically  $\geq 10$  nm long. Importantly, we also observe kinks in the activated behavior of  $R(T)$  that suggest the change of the activation energy as the temperature range is varied. These kinks are attributed to cotunneling processes.

## II. EXPERIMENTAL RESULTS

The fabrication of SWNT devices for four-point measurements has been described in Ref. 15. Briefly,  $\sim 1$ -nm-

diameter SWNT's grown by laser ablation<sup>16</sup> are selected with an atomic force microscopy (AFM). Noninvasive voltage electrodes are defined by positioning two multiwall nanotubes (MWNT's) above the SWNT using AFM manipulation. Cr/Au electrodes are patterned for electric connection using electron-beam lithography [Fig. 1(a)]. The characteristics of the devices are summarized in Table I.

The four-point resistance  $R_{4pt}$  of some SWNT's is particularly large  $> 100$  k $\Omega$  at 300 K. The nature of the scattering centers responsible for this resistance is at present not understood. Figure 1(b) shows the temperature dependence of  $R_{4pt}$  of one of those SWNT's (device 1). The curve is quite flat at high  $T$ , while the resistance increases a lot below 100 K. The high-temperature resistance allows the estimation of the elastic mean free path  $L_e$ . Using  $R_{4pt} = h/4e^2 L/L_e$  with  $L$  the length between the MWNT's,<sup>15</sup> we get  $L_e = 18$  nm.

For comparison, we also show a device that is significantly less resistive at 300 K,  $R_{4pt} = 12$  k $\Omega$ . The  $R_{4pt}(T)$  variation is much less pronounced. This is consistent with previously reported works on two-point, low-Ohmic SWNT devices, where the  $R_{2pt}(T)$  dependence is weak.<sup>17-19</sup> For two-point devices with a large resistance, the resistance has been reported to strongly grow as  $T$  goes to zero, which is usually associated to the change of the contact resistance.<sup>20</sup> In our case, the four-point technique allows to separate the intrinsic and contact resistances.

Figure 1(c) shows that the above measurement in the highly diffusive tube is consistent with an activated behavior of the resistance

$$R_{4pt} = R_0 \exp \frac{E_0}{kT}, \quad (1)$$

with  $E_0$  the activation energy. This dependence is observed in other devices [see Figs. 1(d)–1(f)]. Similar  $R(T)$  behaviors have been reported for disordered wires microfabricated in semiconductors.<sup>21,22</sup>

Figures 1(f)–1(h) show  $R(T)$  measurements on other tubes. Some of them deviate from the standard activated behavior. However, these measurements can be described by successive exponential functions with different activation en-

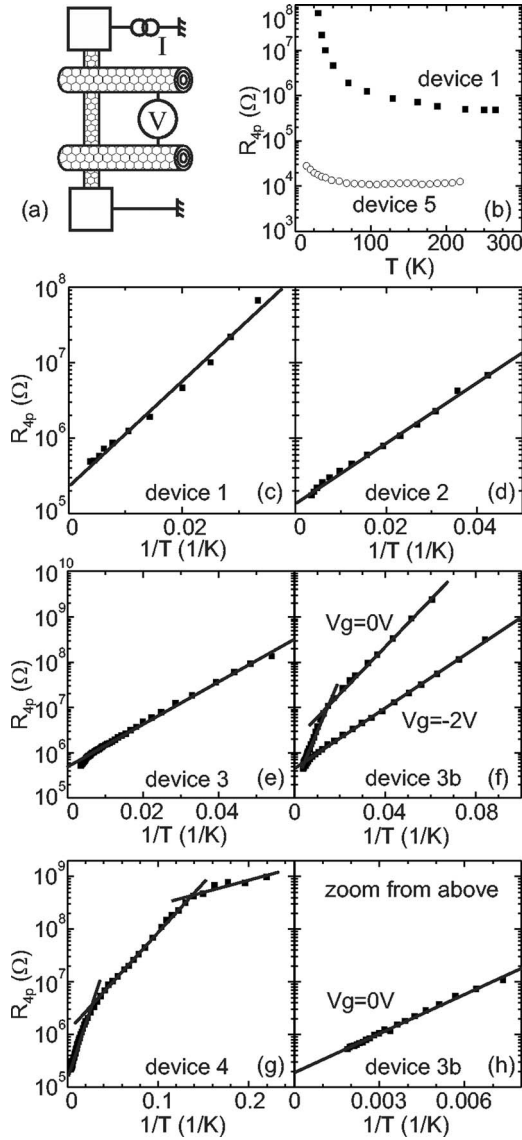


FIG. 1. Four-point resistance. (a) Device schematic. (b)–(h)  $R_{4pt}$  as a function of temperature. When the value of  $V_g$  is not indicated,  $V_g=0$ . Device 3b is the same device as device 3, but it has been measured 1 month before. Microscopic changes might have been occurred in between.

ergies, giving rise to kinks. Interestingly, Fig. 1(f) shows that those two exponential functions can merge in a single one on varying the gate voltage, which is applied on the back side of the Si wafer. Overall, these measurements suggest that the activation energy depends on the temperature range and the gate voltage.

Further insight into transport properties is obtained by studying the high-voltage regime. Figure 2 shows that the differential  $R_{4pt}$  is lowered as  $V_{4pt}$  increases and that the dependence can be fitted with

$$R_{4pt} = R_0 \exp \frac{E_0 - \alpha e V_{4pt}/2}{kT}. \quad (2)$$

This suggests that an increase in the voltage reduces the activation energy. An important point is that the slope de-

TABLE I. Device characteristics.  $L$  is the length between the MWNT's.  $L_{dot}$  is calculated from  $E_0$  extracted at  $V_g=0$ . SC = semiconducting tube with the threshold voltage at  $\sim 40$  V.

	$L$ (nm)	$L_e$ (nm)	$E_0$ (meV)	$L_{dot}$ (nm)	
Device 1	1370	18	13.0	$\sim 50$	SC
Device 2	780	29	7.6	$\sim 90$	Metal
Device 3	1640	20	9.3	$\sim 75$	Metal
Device 4	1000	27	11.5	$\sim 60$	Metal
Device 5	590	300			Metal

duced from Fig. 2 gives  $\alpha$  in Eq. (2) below unity. This means that more than one energy barrier has to be overcome along the tube. A rough estimate of the number of barriers  $N$  can be made by taking  $N=1/\alpha$ , which assumes identical barriers.<sup>21</sup> In this way we obtain  $N \leq 20$ .

Figure 3(a) shows the effect of the gate voltage, which controls the position of the Fermi level in the tube. Large fluctuations of  $R_{4pt}(V_g)$  develop at low  $T$  that look random.<sup>22,23</sup> At first sight, this may question the activation behavior of  $R_{4pt}(T)$  and the kinks discussed above. However, Fig. 3(b) shows that a curve similar to Fig. 1(f) ( $V_g=0$ ) is found by  $V_g$  averaging  $R_{4pt}(T)$ . Moreover, similar dependences are observed, albeit with different activation energies, for the minima and maxima of  $R_{4pt}(V_g)$  as a function of  $T$ . This illustrates the robustness of the activation behavior and the kink for a nanotube with a given Fermi level.

While these fluctuations look random, oscillations can be found that are quite regular within restricted  $V_g$  ranges.<sup>24</sup> Figure 3(c) shows ten successive oscillations. Note that series of regular oscillations can be found at other  $V_g$  ranges and the period is then identical. Interestingly, Figs. 3(d) and 3(e) show that the period can change as the temperature is modified. New oscillations can appear at lower  $T$  that have a shorter period.

### III. DISCUSSION

We now discuss possible origins for the activated behavior of the resistance. One possible mechanism is the Schottky barrier at the interface between a metal electrode and a semiconducting nanotube.<sup>25</sup> However, we also observe the acti-

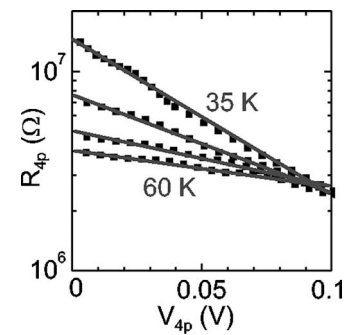


FIG. 2. Four-point differential resistance as a function of  $V_{4pt}$  for device 1 at 35, 45, 55, and 60 K.

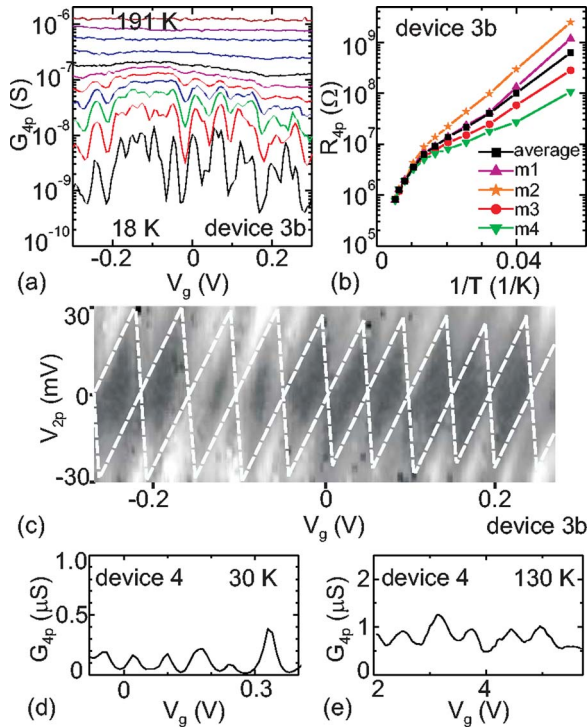


FIG. 3. (Color online) Four-point resistance as a function of the gate voltage. (a)  $R_{4pt}(V_g)$  for device 3b at 18, 25, 31, 39, 49, 60, 74, 96, 125, 158, and 191 K. (b)  $R_{4pt}(T)$  averaged over  $V_g$  between  $-0.3$  and  $0.3$  V and taken at different conductance maxima (m1 at 0.09 V, m2 at 0.17 V, m3 at  $-0.25$  V, m4 at  $-0.05$  V). (c) Two-point differential conductance as a function of  $V_g$  and  $V_{2pt}$  at 20 K. The same measurement with  $G_{4pt}$  is very noisy.  $R_{4pt}(V_g)$  and  $R_{2pt}(V_g)$  in the linear regime show the same features at 20 K. Figures 1(f) and (a) and (c) in this figure are taken in three cooling runs. (d), (e)  $G_{4pt}(V_g)$  for device 4.

vated behavior in metal tubes, which have no Schottky barriers. Moreover, the four-point technique is aimed to avoid contributions from the contacts.<sup>15</sup> Another mechanism is thus needed to account for the results.

The fluctuations of  $R_{4pt}(V_g)$  and the  $R_{4pt}(T)$  dependences may be attributed to universal conductance fluctuations and weak localization. However, the variations of  $R_{4pt}$  are much larger than  $h/e^2$ , so that the results cannot simply originate from interference corrections.

Strong localization (SL) is expected for highly diffusive systems.<sup>10</sup> This theory has been used to explain exponential length dependences of the resistance measured in nanotubes.<sup>26–28</sup> SL occurs when the phase-coherence length  $L_\phi$  becomes longer than the localization length  $L_{loc}$ . This is equivalent to when the width of the coherent states,  $\hbar v_F/L_\phi$ , becomes smaller than the energy separation between the localized states. The localized states are usually regarded as randomly distributed in space and energy [see Fig. 4(a)].<sup>29,30</sup> Irregular oscillations of  $R_{4pt}(V_g)$  are expected, which is in opposition to our results.

We now look at an alternative distribution of localized states as schematized in Fig. 4(b). The tube is here divided into segments separated by highly resistive scattering centers. The segment lengths and therefore the energy separa-

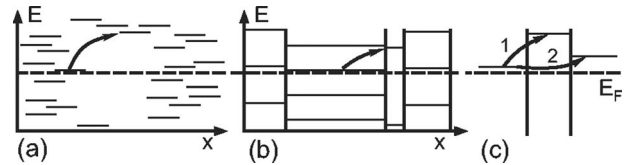


FIG. 4. Schematics of localized states along the nanotube. (a) States are randomly distributed. (b) Strong barriers that define quantum dots. (c) Proposed process to account for the kinks in Figs. 1(f) and 1(g). Arrows represent hopping paths.

tions can be different. At high enough temperatures, levels are thermally smeared out except for the shortest segment that has the largest level separations. Oscillations of  $R_{4pt}(V_g)$  are then regular, and the period is large. At lower temperature, shorter periods arise from longer segments, which agrees with experiments.

So far, the Coulomb interaction between electrons has not been taken into account. However, the charging energy  $E_c$  of a single nanotube quantum dot is known to be larger than the level spacing  $\Delta E$  due to the geometrical confinement of the electron wave.  $\Delta E \approx 0.5$  meV  $\mu\text{m}$ , and the charging energy for a tube dot connected to two tube leads is roughly  $E_c \approx 1.4$  meV  $\mu\text{m}$ .<sup>31</sup> This suggests that the separation in energy between the localized states in Fig. 4(b) is given by the charging energy.

Localization related to a Coulomb blockade through multiple quantum dots<sup>1–4</sup> bears a lot in common with the standard hopping model of the strong localization theory.<sup>22,23,29,30</sup> Series of aperiodic conductance oscillations are expected. Contrary to the SL regime, however, quasiperiodic oscillations are also occasionally predicted, in agreement with experiments. In addition the resistance is expected to be thermally activated, which again agrees with experiments. The activation energy is given by the dot with the level that lies the farthest away from the Fermi level. It may also be the largest separation of energy levels located in neighboring dots. Thus,  $E_0$  is expected to be gate voltage dependent, consistent with our experimental findings.

We here estimate the size of the dots. The activation energy  $E_0$  is roughly  $0.5E_c$  of the shortest dot.  $E_0$  is 11.5 meV for device 4 at high  $T$ . Using  $E_c \approx 1.4$  meV  $\mu\text{m}$ , we get a dot length of  $\sim 60$  nm. Another possibility for this estimation is to use the 625-meV period of the  $R_{4pt}(V_g)$  oscillations at high  $T$  [Fig. 3(e)]. Indeed,  $\Delta V_g \approx 12.5$  meV  $\mu\text{m}$  according to Refs. 31 and 32 for a tube dot connected to two tube leads. This gives  $\sim 20$  nm. Note that  $E_c$  cannot be estimated from the diamond height in Fig. 3(c) since several dots lie in series. Finally, we obtain  $\sim 70$  nm by dividing the tube length by the dot number obtained in Fig. 2. Those three estimations point all to quantum dot lengths of a few 10 nm.

Table I gives the dot length of the other samples, estimated from  $E_0$ . Dot lengths are slightly longer than the elastic length  $L_e$  determined at 300 K.  $L_e$  corresponds to the separation between scatterers when transmissions are 0.5. The barriers that define the quantum dots thus have a transmission  $\leq 0.5$ , which corresponds to a resistance  $\geq 6.5$  k $\Omega$ . This is consistent with the occurrence of a Coulomb blockade since the barrier resistance has to be larger than a few k $\Omega$ 's.

Having shown that the activated behavior of  $R_{4pt}(T)$  originates from a series of quantum dots, we now turn our attention to the kinks [Figs. 1(f)–1(h)]. This may simply come from two thermally activated resistances that lie in series. However, the activation energy would be higher at lower  $T$ , in opposition to the measurements. Another mechanism is needed to describe the kinks.

We propose that the kink is related to a mechanism that is borrowed from the theory of variable-range hopping,<sup>10</sup> see Fig. 4(c). Electrons hop to the neighboring quantum dot as indicated by the arrow 1. At lower  $T$  it pays to make the hop 2 to the second nearest quantum dot. The activation energy is given by the level separation, which is thus reduced. This is in agreement with the experiments.

In the VHR theory such hops are possible thanks to the tunneling process. However, the tunnel probability is here dramatically low since the second nearest dot is a few tens of nanometers far. Another mechanism for the charge transfer between nonadjacent quantum dots is needed to account for the results.

A possible mechanism is that the charge motion between two nonadjacent dots occurs through cotunneling events.<sup>7–9</sup> Cotunneling, which involves the simultaneous tunneling of two or more electrons, transfers the charge via a virtual state. This gives rise to current even when the electron transport is Coulomb blocked.<sup>33</sup> A cotunneling event is called inelastic when the quantum dot is left in an excited state, and the event is otherwise called elastic. For an individual quantum dot contacted to two leads, the conductance contribution of elastic cotunneling is temperature independent, while the contribution of inelastic cotunneling scales as  $T^2$ .

Cotunneling in a series of quantum dots has been recently calculated.<sup>8,9</sup> An energy reservoir supplied by for, e.g., phonons is required since  $\epsilon_i$  the energy of the initial state is most often different than  $\epsilon_f$  the energy of the final state [see hop 2 in Fig. 4(c)]. The resistance contribution between those two states is<sup>8</sup>

$$R \propto R_0^N \exp \frac{\max(|\epsilon_i - \epsilon_f|, |\epsilon_i - \mu|, |\epsilon_f - \mu|)}{kT}, \quad (3)$$

with  $\mu$  the Fermi level and  $N$  the number of dots between the initial and final states.  $R_0 = A_1 E_c / (g \Delta E)$  for elastic cotunneling and  $R_0 = A_2 N^2 E_c^2 / (g(\epsilon_i - \epsilon_f)^2)$  for inelastic cotunneling with  $g = Gh/e^2$  the average dimensionless conductance of a

barrier between two dots and  $A_1$  and  $A_2$  numerical constants of the order of unity. The Coulomb repulsion term between the dots  $i$  and  $f$  is here neglected for simplicity. The prefactor  $R_0^N$  grows as  $N$  the number of involved barriers gets larger. At high temperature, the hopping process between two adjacent dots dominates transport and the prefactor is low [hop 1 in Fig. 4(c)]. As the temperature is reduced, the exponential term grows a lot. It then pays to make the hop between nonadjacent dots when the activation energy is lower [hop 2 in Fig. 4(c)]. This is consistent with the kinks observed in Fig. 1.

The temperature  $T^*$  of the first kink is expected to be around  $kT^* \approx E_0^{above} - E_0^{below}$  with  $E_0^{below}$  and  $E_0^{above}$  the activation energies below and above  $T^*$ . This can be obtained from Eq. (3) taking into account that  $N^{below} - N^{above} = 1$  and that  $\ln R_0$  is of the order of unity. This relation is consistent with the experiments. For example,  $E_0^{above} - E_0^{below} = 14$  meV in Fig. 1(f) for  $V_g = 0$  while  $kT^* = 6$  meV.

We have seen that cotunneling processes allow a slower than thermally activated dependence of the conduction. The main contribution of the conduction comes from one (or a few) quantum dot. The energy levels are randomly positioned in energy, so that we cannot expect a specific functional form for the slower than activated dependence measured here.

#### IV. CONCLUSION

In conclusion, we have shown that the intrinsic resistance of strongly disordered SWNT's is thermally activated. This is due to a Coulomb blockade in a series of  $\geq 10$ -nm-long quantum dots lying along the tube. The activation energy is found to change as the temperature range is changed. We attribute this result to cotunneling processes. Disordered SWNT's form an interesting system for future studies of one-dimensional localization. For example, studies of longer tubes will be investigated to reach the 1D variable-range hopping regime.<sup>34</sup>

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